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Cut-off model and exact general solutions for fragmentation with mass loss

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Abstract. Exact solutions are obtained for a linear rate equation for the evolution of the particle mass distribution during fragmentation with mass loss. These involve general power-law dependences on the particle mass for the fragmentation rate, the daughter-mass distribution, and the mass-loss rate. Consumption of bridges joining two or more otherwise disconnected regions leads to fragmentation such as might occur during the combustion of porous charcoal particles. Exact results for mass-loss rates proportional to the particle mass are relevant to random mass-removal processes such as percolation theory. For pure fragmentation without mass loss, a mass cut-off below which no fragmentation occurs is introduced to avoid the unbounded fragmentation rate for small particles in the 'shattering' regime, in which the fragmentation rate becomes unbounded for particle masses approaching zero. This cut-off model predicts that, for monodisperse initial conditions in the strong shattering regime, the number of particles whose masses exceed the cut-off mass remains near unity for the duration of the fragmentation process, and then drops quickly to zero. The associated asymptotic behaviour excludes the scaling solution in this regime, but includes the scaling solution otherwise.

1. Introduction

The consumption of surface material by an external process such as combustion, oxidation, dissolution, or sublimation can lead to fragmentation of solid porous particles. Under conditions where surface recession occurs within pores as well as at external surfaces, the pores widen and fuse, causing the particle to fall apart eventually as bridges between different regions of the particle are consumed. For highly porous materials, continuous action of the external consumptive process can produce a cascade of fragmentation events. Measurements indicate that hundreds of such fragmentation events can occur during the combustion of a single pulverized coal particle (Dunn-Rankin and Kerstein 1987, 1988, Sarofim *et al* 1977, Sundback *et al* 1985, Quann and Sarofim 1986).

The rate equation approach to fragmentation treats all particles of a particular mass as having the same statistical properties, and offers the prospect of analytical solutions that encompass a range of physical conditions. This approach assigns to each particle a fragmentation rate $a(x)$, which gives the fragmentation probability per unit time for particles of mass x. Ignoring for the moment external consumptive processes and fragmentation of larger particles, the number of such particles decays exponentially with time as $n(x,t) = n_0 e^{-a(x)t}$, with decay constant $a(x)$, just as in radioactive nuclear decay. The rate equation approach thus allows for differences between particles in the time required for fragmentation, thereby reflecting variations in particle shapes and consumption conditions. Even so, all particles of a particular mass share the same statistical properties in this approach; specific

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particle shapes are not considered. Accordingly, we define a distribution $\bar{b}(x|y)$ of daughter particle masses x spawned by the fragmentation of a parent particle of mass y, and a continuous mass-loss rate $c(x)$ giving the mass lost per unit time for particles of mass x. Thus, the approach is best suited for homogeneous combustion and fragmentation conditions, in the absence of fragmentation-inducing interparticle collisions. Such conditions may hold approximately in fluidized bed reactors. The advantage of the rate-equation approach is a physical insight into the time-dependent ensemble particle mass distribution. In contrast with approaches focusing on individual particle shapes, such insight can often be gained through exact or asymptotic analytical solutions.

Whereas numerical simulations typically consider specific particle porosities and consumption conditions (Kerstein and Edwards 1987, Sahimi and Tsotsis 1987, 1988), a linear rate equation encompasses three broad physical regimes (Edwards *et al* 1990, Cai *et al* 1991) capable of describing a spectrum of particle porosities and consumption conditions. These regimes involve the general dimensionless power-law rates $a(x) = x^{\alpha}$, $\bar{b}(x|y) = g(y)x^{\nu}$, and $c(x) = \epsilon x^{\gamma}$. Here the dimensionless parameter $\epsilon \geqslant 0$ measures the overall importance of mass loss relative to fragmentation. The regimes are characterized by the combined exponent $\sigma = \gamma - \alpha - 1$. In the 'fragmentation' regime defined by $\sigma > 0$, relatively high fragmentation rates for small particles allow little mass to be consumed (by combustion, dissolution, etc) between fragmentation events. In the 'recession' regime defined by $\sigma < 0$, relatively low fragmentation rates for small particles allow these particles to be completely consumed without fragmentation. This recession regime is relevant when pores have a minimum separation, the case of primary physical interest. This minimum separation sets the scale for the problem; particles whose sizes are much smaller than this minimum separation will be consumed without fragmentation because of the absence of pores within them. The 'scaling' regime defined by $\sigma = 0$ is the boundary between these two regimes, and exhibits fragmentation behaviour that is independent of mass, that is, small particles behave in the same way as large ones.

We also introduce a cut-off particle mass x_c below which no fragmentation occurs. Every physical system has such a cut-off mass, even if only at the quantum level. Although the recession regime has limited fragmentation for small particles, this cut-off mass is a strict limit below which no fragmentation occurs in any regime. A second motivation for this discrete model is to better understand behaviour in the 'shattering' regime introduced by McGrady and Ziff (1987), for fragmentation with no mass loss $[c(x) = 0]$. The shattering regime is characterized by an unbounded fragmentation rate for small particles, that is, for $\alpha < 0$. In this regime, an infinite number of infinitesimal-mass particles are produced in a finite time. For $\alpha < -1$, the strong shattering regime, this cut-off model surprisingly predicts that, for monodisperse initial conditions, the number of particles whose masses exceed the cut-off mass remains near unity for the duration of the fragmentation process, and then drops quickly to zero. For $\alpha > 0$, cut-off model solutions agree asymptotically with scaling solutions as $t \to \infty$ (Cheng and Redner 1988).

In this paper, section 2 introduces the basic equations and normalization conditions. Section 3 presents explicit solutions for the cut-off model for fragmentation with no mass loss. Section 4 derives exact general solutions with mass loss for arbitrary initial conditions for $\gamma = 1$ (random mass loss) and for $\gamma = -\nu$. To clarify the behaviour in the various regimes, we present specific limits of these solutions for binary fragmentation ($\nu = 0$) and monodisperse initial conditions in section 5. We draw conclusions in section 6.

2. Cut-off model and rate equation

We first introduce the rate equation involving a cut-off particle mass x_c below which no fragmentation occurs. Particles are divided into two states according to their masses $x : x_c < x < \infty$ for the 'fragment state' in which particles can break into smaller particles, and $0 \leqslant x \leqslant x_c$ for the 'dust state' in which no fragmentation occurs.

The linear rate equation

$$\frac{\partial n_f(x,t)}{\partial t} = -a(x)n_f(x,t) + \int_x^\infty a(y)\bar{b}(x|y)n_f(y,t)\,\mathrm{d}y + \frac{\partial}{\partial x}[c(x)n_f(x,t)] \qquad (x > x_c) \tag{1}$$

describes the evolution of the particle mass distribution $n_f(x,t)$ for the fragment state for a continuous system undergoing fragmentation with mass loss. The terms on the right side of equation (1) describe, from left to right, the reduction in the number of particles in the mass range $[x : x + \mathrm{d}x]$ due to the fragmentation of particles in this range, the increase in the number of particles in the range due to fragmentation of larger particles, and the change in the number of particles in the range due to continuous mass loss (Edwards *et al* 1990, Cai *et al* 1991). The external consumptive process ensures that the rate equation is linear: fragmentation due to repeated collisions between particles requires a nonlinear rate equation (Cheng and Redner 1988, 1990). The evolution of the particle mass distribution $n_d(x,t)$ for the dust state is correspondingly governed by

$$\frac{\partial n_d(x,t)}{\partial t} = \int_{x_c}^\infty a(y)\bar{b}(x|y)n_f(y,t)\,\mathrm{d}y \qquad (x \leqslant x_c) \tag{2}$$

since particles in this state can neither break nor lose mass.

A normalization condition for $\bar{b}(x|y)$,

$$y = \int_0^y x\bar{b}(x|y)\,\mathrm{d}x \qquad (y > x_c) \tag{3}$$

demands mass conservation during fragmentation events, namely, it requires that the mass y of a parent particle just before it breaks equals the sum $\int_0^y x\bar{b}(x|y)\,\mathrm{d}x$ of the masses of the resulting daughter particles. A useful relation for the time rate of change of total mass $M = M_f + M_d = \int_{x_c}^\infty xn_f(x,t)\,\mathrm{d}x + \int_0^{x_c} xn_d(x,t)\,\mathrm{d}x$ follows by multiplying equations (1) and (2) by x and by integrating;

$$\mathrm{d}M/\mathrm{d}t = -\int_{x_c}^\infty c(x)n_f(x,t)\,\mathrm{d}x. \tag{4}$$

In the absence of mass loss $[c(x) = 0]$, the total particle mass M is evidently conserved.

For the power-law rates introduced earlier, the normalization condition, (3), implies that $g(y) = (\nu+2)/y^{\nu+1}$ with $\nu > -2$, so that (1) and (2) become

$$\frac{\partial n_f(x,t)}{\partial t} = -x^\alpha n_f(x,t) + (\nu+2)x^\nu \int_x^\infty y^{\alpha-\nu-1}n_f(y,t)\,\mathrm{d}y + \epsilon\frac{\partial}{\partial x}[x^\gamma n_f(x,t)] \tag{5}$$

and

$$\frac{\partial n_d(x,t)}{\partial t} = (\nu+2)x^\nu \int_{x_c}^\infty y^{\alpha-\nu-1}n_f(y,t)\,\mathrm{d}y. \tag{6}$$

The ensemble-averaged number of daughter particles produced by a fragmentation event is given by $\bar{\mathcal{N}} = \int_0^y \bar{b}(x|y)\,\mathrm{d}x = (\nu+2)/(\nu+1)$ for $\nu > -1$, whereas $\bar{\mathcal{N}} = \infty$ for $-2 < \nu \leqslant -1$ (McGrady and Ziff 1987). The requirement $\bar{\mathcal{N}} \geqslant 2$ that fragmentation events produce two or more fragments immediately yields the upper limit of the allowable range $-1 < \nu \leqslant 0$.

3. Explicit solutions for no mass loss with $\epsilon = 0$

In the absence of mass loss ($\epsilon = 0$), equations (5) and (6) become

$$\frac{\partial n_f(x,t)}{\partial t} = -x^\alpha n_f(x,t) + (2+\nu)x^\nu \int_x^\infty y^{\alpha-\nu-1} n_f(y,t)\,\mathrm{d}y \tag{7}$$

and

$$\frac{\partial n_d(x,t)}{\partial t} = (2+\nu)x^\nu \int_{x_c}^\infty y^{\alpha-\nu-1} n_f(y,t)\,\mathrm{d}y. \tag{8}$$

The general solution of equation (7) has been obtained by using the Laplace transform approach (Huang *et al* 1991). Here, we generalize these solutions to include a mass cut-off. For arbitrary initial conditions $n_f(x,0)$ and $n_d(x,0)$, the general solution of equation (7) is (Huang *et al* 1991)

$$n_f(x,t) = \mathrm{e}^{-x^\alpha t}\left\{ n_f(x,0) + m\alpha t x^\nu \int_x^\infty y^{\alpha-\nu-1} n_f(y,0) F_1[1-m, 2, t(x^\alpha - y^\alpha)]\,\mathrm{d}y \right\}. \tag{9}$$

The general solution for $n_d(x,t)$ follows by integrating equation (8);

$$n_d(x,t) = n_d(x,0) + m\alpha x^\nu \int_{x_c}^\infty y^{\alpha-\nu-1}\,\mathrm{d}y \int_0^t n_f(y,t')\,\mathrm{d}t'. \tag{10}$$

Here, $F_1(a,b,x)$ is the confluent hypergeometric function and $m = (2+\nu)/\alpha$.

For monodisperse initial conditions $n_f(x,0) = \delta(x-l)$ and $n_d(x,0) = 0$, we use Kummer's identity $F_1(a,b,x) = \mathrm{e}^x F_1(b-a, b, -x)$ to simplify (9) to

$$n_f(x,t) = \mathrm{e}^{-l^\alpha t}\{\delta(x-l) + (2+\nu)t l^{\alpha-\nu-1} x^\nu F_1[1+m, 2, t(l^\alpha - x^\alpha)]\}. \tag{11}$$

Equation (11) describes the evolution of particles in the fragment state. Eventually, all particles in the fragment state will break into dust particles, and then the fragmentation process ceases. To find the final total number $N_d(\infty)$ of dust particles, we substitute (11) into (10) and integrate over $[0, x_c]$ to find $N_d(\infty) = \bar{\mathcal{N}} l/x_c$, where $\bar{\mathcal{N}} = (\nu+2)/(\nu+1)$ is the ensemble-averaged number of daughter particles produced by one fragmentation event. Thus, the final total number of dust particles is the product of the ensemble-averaged number $\bar{\mathcal{N}}$ of daughter particles produced by one fragmentation event and the average number l/x_c of fragmentation events occurring in the whole fragmentation process. This result is independent of the details of the fragmentation process and applies to any values of α and ν

The time rate of change of the total mass $M_f(t)$ $[M_d(t)]$ in the fragment (dust) state is

$$\frac{\mathrm{d}M_f(t)}{\mathrm{d}t} = -\frac{\mathrm{d}M_d(t)}{\mathrm{d}t} = -\mathrm{e}^{-l^\alpha t} l^{\alpha-\nu-1} x_c^{\nu+2} F_1[m, 1, t(l^\alpha - x_c^\alpha)] \leqslant 0. \tag{12}$$

The monotonically decreasing (increasing) behaviour of $M_f(t)$ $[M_d(t)]$ reflects the fact that any production of dust particles is accompanied by a mass flow from the fragment state to the dust state. The vanishing sum of these two rates ensures mass conservation for any value of α, so that

$$M_f(t) + M_d(t) = M_f(0) + M_d(0) = l. \tag{13}$$

Equation (13) resolves a difficulty encountered in the previous continuous fragmentation model (McGrady and Ziff 1987, Huang *et al* 1991), where the explicit solutions for $\alpha < 0$ yield a decreasing total mass in the whole system and therefore violate mass conservation.

It is instructive to study the asymptotic behaviour of (11) for very large t;

$$n_f(x,t) \to \begin{cases} \dfrac{\alpha l}{\Gamma(m)} t^{2/\alpha} \exp(-x^\alpha t)(x^\alpha t)^{\nu/\alpha} & \alpha > 0 \\ -\dfrac{\alpha l^{\alpha-\nu-1}}{\Gamma(-m)} t^{-m} \exp(-l^\alpha t) x^{-(\alpha+2)} & \alpha < 0. \end{cases} \tag{14}$$

This asymptotic behaviour has the scaling form (Cheng and Redner 1988)

$$n(x,t) = t^{2/\alpha} f(xt^{1/\alpha}) \tag{15}$$

for $\alpha > 0$, but does not for $\alpha < 0$. This result reflects the lack of scale invariance in the shattering regime.

For integer m, (9) can be expressed using associated Laguerre polynomials (Huang *et al* 1991). These explicit polynomial solutions are helpful in investigating the nature of the fragmentation processes. Here, we choose some simple representative cases.

(i) For $m = -1$ ($\alpha = -\nu - 2 < 0$) with monodisperse initial conditions $n_f(x,0) = \delta(x-l)$ and $n_d(x) = 0$, the explicit solutions are

$$n_f(x,t) = \mathrm{e}^{-l^\alpha t}[\delta(x-l) + (\nu+2) l^{\alpha-\nu-1} t x^\nu] \tag{16}$$

$$n_d(x,t) = (\nu+2) l^{\alpha+1}[(1-\mathrm{e}^{-l^\alpha t}) r_c^\alpha + (1-r_c^\alpha) t l^\alpha \mathrm{e}^{-l^\alpha t}] x^\nu \tag{17}$$

where $r_c = x_c/l$. The total numbers and masses of particles in the two states are

$$N_f(t) = \mathrm{e}^{-l^\alpha t}[1 + \bar{\mathcal{N}} t l^\alpha (1 - r_c^{\nu+1})] \tag{18}$$

$$N_d(t) = \bar{\mathcal{N}}[(1-\mathrm{e}^{-l^\alpha t})/r_c + (1-r_c^\alpha) t l^\alpha r_c^{\nu+1} \mathrm{e}^{-l^\alpha t}] \tag{19}$$

$$M_f(t) = l\mathrm{e}^{-l^\alpha t}[1 + t l^\alpha (1 - r_c^{\nu+2})] \tag{20}$$

$$M_d(t) = l[1 - \mathrm{e}^{-l^\alpha t} - (1 - r_c^{\nu+2}) t l^\alpha \mathrm{e}^{-l^\alpha t}]. \tag{21}$$

These solutions obviously satisfy the initial conditions $N_f(0) = 1$, $N_d(0) = 0$, $M_f(0) = l$, and $M_d(0) = 0$. The limiting behaviour is $N_f(\infty) = 0$, $N_d(\infty) = \bar{\mathcal{N}} l/x_c$, $M_f(\infty) = 0$, and $M_d(\infty) = l$, indicating that all particles in the fragment state eventually break into dust particles, after which the break-up process ceases as expected. The number of particles in the fragment state $N_f(t)$ increases initially from unity, reaches a peak, and eventually decreases to zero as all particles break into dust particles. To examine this peak, we take the derivative of (18) and set $\mathrm{d}N_f(t)/\mathrm{d}t = 0$, yielding $t_c \approx 1/(\nu+2) l^\alpha$ for $r_c \ll 1$. Substituting t_c into (18), we find the maximum total number of particles in the fragment state,

$$N_f(t_c) \approx \bar{\mathcal{N}} \mathrm{e}^{-1/(\nu+2)}. \tag{22}$$

For binary break-up ($\nu = 0$) with $\alpha = -2$, (22) implies that the average number of fragment particles never exceeds $N_f(t_c) \approx 1.21$. This result is valid for $r_c \ll 1$ and is *independent* of x_c in this limit. Thus, for $x_c \to 0$, the number of finite-mass particles, averaged over many individual realizations, never appreciably exceeds unity. This small average number is surprising because an infinite number of fragmentation events are required to produce an infinite number of infinitesimal-mass 'dust' particles for $x_c \to 0$; how could the average number of finite-mass particles remain so low during this process? The answer lies in the fragmentation rate $a(x) = x^\alpha = x^{-2}$ for particles of mass x. For this fragmentation rate, the average lifetime of a particle before fragmentation goes as the square of the particle mass; $a(x)^{-1} = x^2$. Hence, very small particles break extremely quickly. After a fragmentation event, the small daughter typically breaks quickly compared with the large daughter, and the daughters of the small daughter break even more quickly, and so on. Thus, the time required for a small daughter and all of her descendants to disappear into dust, during which the number of finite-mass particles is large, is typically small compared with the lifetime of the large daughter. Hence, the fragmentation process favours a state in which there is one large particle waiting to break, during which the average number of particles is unity. To better understand the small average numbers of particles, we studied a discrete model (Edwards and Huang) for binary fragmentation in which each fragmentation event occurs at the average particle lifetime $a(x)^{-1}$ and produces daughters of average mass (one daughter of mass $y/4$ and the other of mass $3y/4$ for a parent of mass y). This model confirms that the small average number of finite-mass particles is a result of the short time taken by a small daughter and all of her descendants to disappear into dust, compared with the lifetime of the large daughter. For non-binary break-up ($-1 < \nu < 0$), (22) implies that the peak $N_f(t_c)$ in the average number of particles in the fragment state is even lower than the number for binary break-up.

(ii) For $\nu = 0$ and $\alpha = -1$ ($m = -2$) with monodisperse initial conditions $n_f(x, 0) = \delta(x - l)$ and $n_d(x) = 0$, the explicit solutions are

$$n_f(x,t) = \mathrm{e}^{-l^{-1}t}[\delta(x-l) + 2l^{-2}t(1-(l^{-1}-x^{-1})t/2)] \tag{23}$$

$$n_d(x,t) = \frac{2}{lr_c^2}\{1 - [1 + (1-r_c^2)tl^{-1} + \tfrac{1}{2}(1-r_c)^2t^2l^{-2}]\mathrm{e}^{-l^{-1}t}\}. \tag{24}$$

The total numbers and masses of particles in the two states are

$$N_f(t) = \mathrm{e}^{-l^{-1}t}[1 + 2(1-r_c)tl^{-1} + t^2l^{-2}(r_c - \ln r_c - 1)] \tag{25}$$

$$N_d(t) = \frac{2}{r_c}\{1 - [1 + (1-r_c^2)tl^{-1} + \tfrac{1}{2}(1-r_c)^2t^2l^{-2}]\mathrm{e}^{-l^{-1}t}\} \tag{26}$$

$$M_f(t) = l\mathrm{e}^{-l^{-1}t}[1 + (1-r_c^2)tl^{-1} + \tfrac{1}{2}t^2l^{-2}(1-r_c)^2] \tag{27}$$

$$M_d(t) = l\{1 - [1 + (1-r_c^2)tl^{-1} + \tfrac{1}{2}t^2l^{-2}(1-r_c)^2]\mathrm{e}^{-l^{-1}t}\}. \tag{28}$$

Taking the limit $r_c \to 0$ ($x_c \to 0$) in equation (25) yields an infinite number $N_f(t)$ of particles in the fragment state for $t > 0$. Since the cut-off mass x_c always has a finite value, we see that the cut-off model resolves this divergence problem. Taking the derivative of (25) and setting $\mathrm{d}N_f(t)/\mathrm{d}t = 0$ yield $t_c \approx 2l$ for $r_c \ll 1$. Substituting t_c into (25), we obtain a maximum total number of particles in the fragment state which depends on r_c;

$$N_f(t_c) \approx \mathrm{e}^{-2}(5 - 4\ln r_c). \tag{29}$$

For $r_c = 10^{-4}$, (29) yields the maximum total number of particles in the fragment state, $N_f(t_c) \approx 6$. This particular case was chosen in previous work (Ziff *et al*, Huang *et al* 1991) to illustrate the nature of the 'shattering' transition in fragmentation. Solving (25) and (26) numerically as a function of $l^{-1}t$ illustrates the evolution of the total particle numbers in the two states (figure 1).

These two cases illustrate the general behaviour of the fragmentation processes for $\alpha < 0$. For $\alpha = -2$, the total number of particles in the fragment state increases slowly from unity, reaches a peak at $N_f(t_c) = 1.21$ that is independent of r_c, and then decreases to zero. The total number of particles in the dust state increases dramatically during this time, reflecting the high fragmentation rates $a(x) = x^{-2}$ for small particles. The smaller the particle mass, the larger the fragmentation rate and the faster it breaks into daughters. For $\alpha = -1$, the fragmentation rate $a(x) = x^{-1}$ for small particles is smaller than for $\alpha = -2$. This results in a peak number $N_f(t_c)$ that depends on r_c (29), and that becomes infinite as $r_c \to 0$. More generally, integrating equation (14) indicates that, for $\alpha < -1$, the total

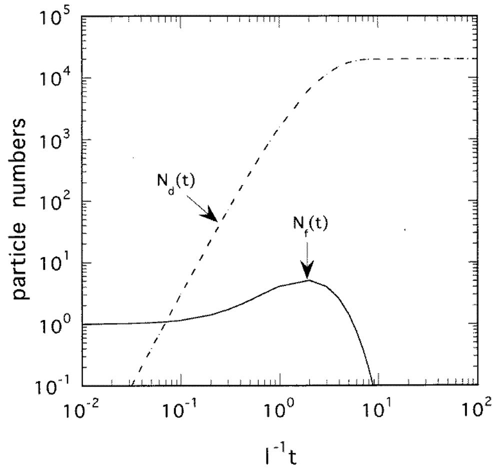

Figure 1. The evolution of the total particle number $N_f(t)$ $[N_d(t)]$ in the fragment (dust) state as a function of $l^{-1}t$, (25) and (26), for $\alpha = -1$ with monodisperse initial conditions, binary break-up, and $l/x_c = 10^4$.

number of finite-mass particles remains finite during the entire fragmentation process, and is independent of r_c for $r_c \ll 1$. This is called the strong shattering regime because of the rapid production of dust particles. For $-1 \leqslant \alpha < 0$, the total number of particles in the fragment state depends on r_c, and becomes unbounded as $r_c \to 0$. This dependence on r_c implies that small particles with masses near x_c have large enough lifetimes to play a significant role in the overall particle average. By contrast, in the strong shattering regime, such particles shatter too quickly to be significant.

(iii) We now consider $\alpha > 0$. For $\alpha = \nu + 2$ $(m = 1)$ with monodisperse initial conditions $n_f(x, 0) = \delta(x - l)$ and $n_d(x) = 0$, the explicit solutions are

$$n_f(x, t) = \mathrm{e}^{-x^\alpha t}[\delta(x - l) + (\nu + 2)tlx^\nu] \tag{30}$$

$$n_d(x, t) = (\nu + 2)lx_c^{-\alpha}(1 - \mathrm{e}^{-x_c^\alpha t})x^\nu. \tag{31}$$

The total numbers of particles in the two states are

$$N_f(t) = \mathrm{e}^{-l^\alpha t} + (\nu + 2)lt \int_{x_c}^{l} x^\nu \mathrm{e}^{-x^\alpha t}\, \mathrm{d}x \tag{32}$$

$$N_d(t) = \bar{\mathcal{N}} l x_c^{-1}(1 - \mathrm{e}^{-x_c^\alpha t}). \tag{33}$$

For $\nu = 0$ $(\alpha = 2)$ and $l/x_c = 10^4$, solving (32) and (33) numerically as a function of $x_c^2 t$ illustrates the evolution of the total particle numbers in the two states (figure 2). Contrary to the fragmentation for $\alpha < 0$, the number of particles in the fragment state overwhelms the number of particles in the dust state through all but the last stages of fragmentation. This shows that fragmentation is mainly dominated by events in which almost all daughter particles are still in the fragment state and continue to break into smaller particles. Finally, when almost all particles in the fragment state have masses comparable with the cut-off mass, they break into dust particles as indicated in figure 2, reflecting the fact that the smaller the particle mass, the smaller the fragmentation rate.

The total masses in the two states $M_f(t) = l\mathrm{e}^{-x_c^\alpha t}$ and $M_d(t) = l(1 - \mathrm{e}^{-x_c^\alpha t})$ vary exponentially with time t with a dimensionless time constant $\tau = x_c^{-\alpha}$, which indicates that

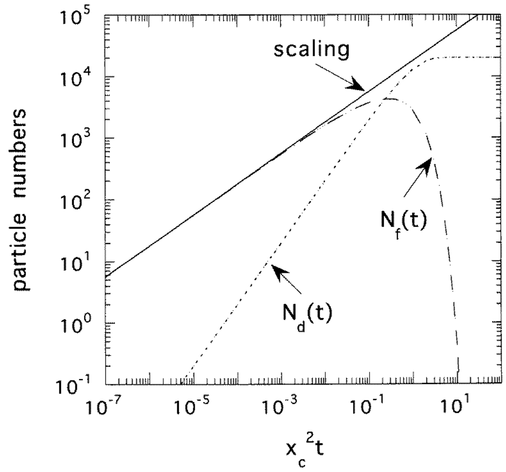

Figure 2. The evolution of the total particle number $N_f(t)$ $[N_d(t)]$ in the fragment (dust) state as a function of $x_c^2 t$, (32) and (33), for $\alpha = 2$ with monodisperse initial conditions, binary break-up, and $l/x_c = 10^4$. The scaling solution is from (34).

the smaller the cut-off mass, the larger the time constant, and therefore the longer the time for completion of fragmentation.

After the initial break-up, the second term in (30) dominates the solution, and (30) thereby reduces to the scaling form of (15). The total number of particles in the fragment state has the approximate relation,

$$N_f(t) = (\nu + 2) l t^{1/\alpha} \int_0^\infty \xi^\nu \mathrm{e}^{-\xi^\alpha} \, \mathrm{d}\xi \tag{34}$$

where $\xi = x t^{1/\alpha}$. For binary break-up, (34) reduces to $N_f(t) = \sqrt{\pi} l t^{1/2}$, which agrees well with (32) initially and before almost all particles in the fragment state break into dust particles (see figure 2).

4. General solutions for fragmentation with mass loss

The value of γ reflects the nature of the mass removal process, the extent of pore penetration, and the particle morphology. For combustion of porous particles, the extent of pore penetration is governed by the temperature. At low temperatures barely sufficient to support very slow combustion, oxygen is plentiful at the particle surface and within the pores, allowing combustion both at the external surface and within the pores, with an overall particle mass-loss rate $c(x) = \epsilon x^\gamma \sim S$ proportional to the overall particle surface area S (including the area of the pore walls) in the limit of extremely slow combustion. Hence, at these low temperatures, the chemical kinetics limits the surface recession. For a wide range of fractal and compact objects, the surface area is related to the mass according to a power law $S \sim x^\gamma$, with $\gamma = \frac{2}{3}$ for solid spheres.

At high temperatures with high combustion rates, oxygen reaching the particle is immediately consumed at the external surface of the particle, with no opportunity to diffuse into the pores. Hence, at these high temperatures, oxygen diffusion limits the surface recession, and the overall mass-loss rate follows by solving Laplace's equation for the oxygen concentration with zero concentration at the particle surface and an oxygen flux per unit surface area proportional to the normal component of the concentration gradient at the surface, yielding $\gamma = \frac{1}{3}$ for solid spheres.

The value $\gamma = 0$ implies a mass-loss rate that is independent of the particle mass, relevant for mass removal from the ends of a quasi-one-dimensional object. This value can be thought of as the extreme limit of diffusion-limited combustion, where only the very tips of a tenuous object are consumed. The only known solution with continuous mass-loss and power-law rates is for $\gamma = 0$ and $\alpha = 1$ (Huang *et al* 1991).

Here, we present a solution for $\gamma = -\nu$ with $0 \leqslant \gamma < 1$ and $\alpha = 1+\nu$ with $0 < \alpha \leqslant 1$, and a solution for $\gamma = 1$ and arbitrary α. The solution for arbitrary α allows us to explore the various regimes by changing the value of α. The value $\gamma = 1$ implies a mass-loss rate that is proportional to the particle mass. This means that the probability of removal of an element of mass from a particle is independent of the location of the mass element within the particle, allowing mass removal from the external surfaces, from the pore walls, and from the deep interior with equal probability. Thus, the value $\gamma = 1$ for random mass removal can be thought of as an extreme limit of chemical-kinetics-limited combustion. This value is also relevant to percolation theory; removing occupied bonds (or sites) at random on a lattice yields a mass-loss rate proportional to the cluster mass. Furthermore, since the surface area or 'perimeter' of critical percolation clusters is proportional to the cluster mass, $\gamma = 1$ for removal from the surface as well.

To obtain solutions with continuous mass loss, it is convenient to transform (5) according to $x = u^{1/\alpha}$ and $n(x,t) = x^\nu w(u,t)$;

$$\frac{\partial w_f(u,t)}{\partial t} = -u w_f(u,t) + m \int_u^\infty w_f(v,t)\,\mathrm{d}v + \eta u^{\mu-1} w_f(u,t) + \beta u^\mu \frac{\partial w_f(u,t)}{\partial u} \tag{35}$$

where $m = (\nu+2)/\alpha$, $\eta = \epsilon(\gamma+\nu)$, $\mu = (\gamma+\alpha-1)/\alpha$, and $\beta = \epsilon\alpha$.

(i) Setting $\eta = 0$ and $\mu = 0$ yields $\gamma = -\nu$ with $0 \leqslant \gamma < 1$ and $\alpha = 1+\nu$ with $0 < \alpha \leqslant 1$. Equation (35) yields

$$\frac{\partial w_f(u,t)}{\partial t} = -u w_f(u,t) + m \int_u^\infty w_f(v,t)\mathrm{d}v + \beta \frac{\partial w_f(u,t)}{\partial u}. \tag{36}$$

The trial transformations $\xi = \xi(u,t)$, $\tau = \tau(u,t)$, and $w_f(u,t) = F(t)G(\xi,\tau)$ give

$$\frac{\partial w_f(u,t)}{\partial t} = \frac{\mathrm{d}F(t)}{\mathrm{d}t} G(\xi,\tau) + F(t)\left[\frac{\partial G(\xi,\tau)}{\partial \xi}\frac{\partial \xi}{\partial t} + \frac{\partial G(\xi,\tau)}{\partial \tau}\frac{\partial \tau}{\partial t}\right]$$

$$\beta \frac{\partial w_f(u,t)}{\partial u} = F(t)\left[\beta \frac{\partial G(\xi,\tau)}{\partial \xi}\frac{\partial \xi}{\partial u} + \beta \frac{\partial G(\xi,\tau)}{\partial \tau}\frac{\partial \tau}{\partial u}\right].$$

Setting $\frac{\partial \tau}{\partial u} = 0$ and $\frac{\partial \xi}{\partial t} = \beta \frac{\partial \xi}{\partial u}$ yields $\tau = \tau(t)$ and $\xi = u + \beta t$. Thus, (36) reduces to

$$\frac{\mathrm{d}F(t)}{\mathrm{d}t} G(\xi,\tau) + F(t)\frac{\partial G(\xi,\tau)}{\partial \tau}\frac{\mathrm{d}\tau}{\mathrm{d}t} = -(\xi - \beta t)F(t)G(\xi,\tau) + mF(t)\int_\xi^\infty G(\zeta,\tau)\,\mathrm{d}\zeta.$$

Setting $\frac{\mathrm{d}\tau}{\mathrm{d}t} = 1$ and $\frac{\mathrm{d}F(t)}{\mathrm{d}t} = \beta t F(t)$ yields $\tau = t$ and $F(t) = \mathrm{e}^{\beta t^2/2}$. Substituting $\tau = t$, $\xi = u + \beta t$, and $w_f(u,t) = \mathrm{e}^{\beta t^2/2} G(\xi,\tau)$ into (36), we have

$$\frac{\partial G(\xi,t)}{\partial t} = -\xi G(\xi,t) + m \int_\xi^\infty G(\zeta,t)\,\mathrm{d}\zeta. \tag{37}$$

The general solution of (37) has been obtained using a Laplace transform approach (Huang *et al* 1991),

$$G(\xi,t) = \mathrm{e}^{-\xi t}[G(\xi,0) + mt \int_\xi^\infty G(\zeta,0) F_1[1-m, 2, t(\xi-\zeta)]\,\mathrm{d}\zeta]. \tag{38}$$

Written using the original physical variables, equation (38) becomes

$$n_f(x,t) = x^\nu e^{\beta t^2/2} e^{-\xi t} \Big\{ \xi^{-\nu/\alpha} n_f(\xi^{1/\alpha}, 0) + mt \int_\xi^\infty \zeta^{-\nu/\alpha} n_f(\zeta^{1/\alpha}, 0) F_1[1-m, 2, t(\xi-\zeta)]\, d\zeta \Big\} \quad (39)$$

where $\xi = x^\alpha + \beta t$.

(ii) Setting $\mu = 1$ gives $\gamma = 1$ and $-\infty < \alpha < \infty$. Equation (35) becomes

$$\frac{\partial w_f(u,t)}{\partial t} = -(u-\eta) w_f(u,t) + m \int_u^\infty w_f(v,t)\, dv + \beta u \frac{\partial w_f(u,t)}{\partial u}. \quad (40)$$

Using the same technique adopted in the previous case, we find the appropriate transformation $\xi = u e^{\beta t}$, $\tau = (1 - e^{-\beta t})/\beta$, and $w_f(u,t) = e^{\eta t} G(\xi, \tau)$. Under this transformation, (40) reduces to (37) by substituting τ for t. The general solution of $G(\xi, \tau)$ has the same form of (38) except for this substitution. We can write the general solution in the original physical variables,

$$n_f(x,t) = e^{\epsilon t - \tau' x^\alpha} \Big\{ n_f(x e^{\epsilon t}, 0) + (\nu+2)\tau' x^\nu \int_x^\infty y^{\alpha-\nu-1} n_f(y e^{\epsilon t}, 0) F_1[1-m, 2, \tau'(x^\alpha - y^\alpha)]\, dy \Big\} \quad (41)$$

where $\tau' = \tau e^{\beta t}$.

5. Monodisperse initial condition for fragmentation with mass loss

It is instructive to consider the monodisperse initial condition $n(x,0) = \delta(x-l)$. We choose two representative cases to illustrate the behaviour of fragmentation with mass loss.

(i) For $\nu = 0$, $\alpha = 1$, and $\gamma = 0$, (39) becomes

$$n_f(x,t) = \exp(-xt - \epsilon t^2/2)[\delta(x - x_t) + 2t + x_t t^2 - x t^2]. \quad (42)$$

The total number of particles and the total mass of the particles in the fragment state are

$$N_f(t) = \exp(-\epsilon t^2/2 - x_c t)(1 + x_t t - x_c t) \quad (43)$$

$$M_f(t) = \exp(-\epsilon t^2/2 - x_c t)(x_t + x_t x_c t - x_c^2 t) \quad (44)$$

where $x_t = l - \epsilon t$ and $x_c \leqslant x \leqslant x_t$.

(ii) For $\nu = 0$, $\alpha = 1$, and $\gamma = 1$, (41) becomes

$$n_f(x,t) = \exp(-x\tau')[\delta(x - x_t) + 2\tau' + x_t \tau'^2 - x\tau'^2]. \quad (45)$$

The total number of particles and the total mass of the particles in the fragment state are

$$N_f(t) = \exp(-x_c \tau')(1 + x_t \tau' - x_c \tau') \quad (46)$$

$$M_f(t) = \exp(-x_c \tau')(x_t + x_t x_c \tau' - x_c^2 \tau') \quad (47)$$

where $x_t = l e^{-\epsilon t}$, $\tau' = (e^{\epsilon t} - 1)/\epsilon$, and $x_c \leqslant x \leqslant x_t$.

The first terms in (42) and (45) account for unbroken particles of mass $x = x_t = l - \epsilon t$ for the first case and $x = x_t = l \exp(-\epsilon t)$ for the second case. In each case, the unbroken particles have a mass x_t which decreases monotonically with time due to continuous mass loss and which satisfies $c(x_t) = -dx_t/dt$. The rest of the terms in these equations account for smaller broken particles. Such terms vanish at $t = \tau' = 0$ but clearly dominate at large times.

The constant continuous mass-loss rate $c(x) = \epsilon$ for $\gamma = 0$ implies that the mass loss per unit time for a particle is independent of its mass. For $\alpha = 1$, the smaller the particle mass, the smaller the particle fragmentation rate. Thus, small particles typically lose all of their mass without breaking, reflecting the behaviour in the strong-recession regime ($\sigma = \gamma - \alpha - 1 = -2$). For $\gamma = 1$, both the continuous mass-loss rate $c(x) = \epsilon x$ and the fragmentation rate $a(x) = x$ are proportional to the particle mass. Hence, the smaller the particle mass, the smaller the fragmentation rate and the continuous mass-loss rate, implying the weak-recession regime ($\sigma = -1$). A numerical calculation based on (43) and (46) with $l = 1.0$, $\epsilon = 10^{-3}$, and $x_c = 10^{-6}$ yields the maximum total number of particles ≈ 14 for $\gamma = 0$ and ≈ 94 for $\gamma = 1$. The fragmentation process for $\gamma = 1$ lasts 100 times longer than that for $\gamma = 0$.

6. Conclusions

A cut-off model avoids the unbounded fragmentation rate for small particles in the 'shattering' regime, and provides a model to study physical systems involving particles that cease to break when their masses fall below a lower limit. Exact solutions are presented for this model for mass-conserving fragmentation. These solutions show that the parameter α in the fragmentation rate characterizes these mass-conserving fragmentation processes. The asymptotic behaviours of these solutions indicate that the scaling solution dominates the fragmentation for $\alpha > 0$, but is prohibited for $\alpha < 0$. This cut-off model reveals that, for monodisperse initial conditions with $\alpha < -1$, the number of particles whose masses exceed the cut-off mass remains near unity for the duration of the fragmentation process, and then drops quickly to zero.

Exact general solutions for fragmentation with mass loss for arbitrary initial conditions for $\gamma = 1$ (random mass loss) and for $\gamma = -\nu$ have also been presented. Mass-loss rates proportional to the particle mass are relevant to random mass-removal processes such as percolation theory for which the probability of consumption of a mass element is independent of the location of the mass element and of the imbedding dimension, and depends only on the cluster mass. The solutions for binary fragmentation and monodisperse initial conditions show that, in the recession regime, low fragmentation rates for small particles allow these particles to be completely consumed without fragmentation.

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